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LABS INC MURRAY HILL NJ P M RENTZEPIS 1983

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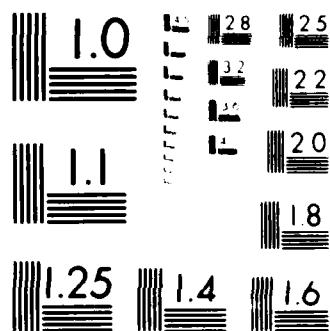
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Time Resolved Resonance Raman Conference Royal
Institution, London U. K.

P. M. Rentzepis

Bell Laboratories
Murray Hill, New Jersey 07974

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1983

The purpose of the conference was to bring together a group of approximately 60 internationally known experts in the field of time resolved Raman spectroscopy to present and discuss their most recent work, to an audience of ~90 scientists. The presentations in the form of 45 min lectures and long discussion was focussed on the advances made in time resolved Raman spectroscopy, the inherent difficulties which have hindered a rapid development of this field and the most fruitful experimental paths to be followed for further advances.

Most if not all of the time resolved Raman experiments are restricted to a very large extent to nanosecond and longer time regime except for three or four laboratories in the United States which are in the process of establishing picosecond time resolved Raman systems. These include [picosecond Raman] the laboratories of Prof's M. A. El Sayed, UCLA L. Styer Stanford and P. M. Rentzepis Bell Labs.

The experimental difficulties in time resolved Raman arise from very low signals, compared to emission processes such as fluorescence, attributable mostly to:

1. Low Raman crosssection.

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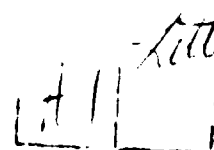
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2. Raman lines from the solvent or the host crystal which is usually at a concentration of $\sim 10^4$ higher than the solute studied.
3. The energy per pulse, in the picosecond experiments is in the micro-joule range therefore the Raman signal intensity is extremely weak and of need to average the signals for long periods of time.
4. Means for rejection of fluorescence before its entrance into the spectrometer and/or detector.

All these aspects were discussed in great detail and consumed the largest portion of the meeting which had a duration of 8 hrs.

Item 1. weak crosssection is a natural limitation of Raman scattering processes and there no means for increasing it in spontaneous Raman. However, it is possible to increase the scattering intensity by 10^3 or more by selecting the scattering pulse wavelength to be near an absorption resonance level. Such process known as resonance Raman is employed by practically all time resolved Raman practitioners. In fact the time resolved data presented were all performed in the resonance mode. Item (2) the solvent Raman lines cannot be eliminated, however, their effect may be minimized by selecting a solvent which does not have Raman emission in the spectral region of the solute



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or at least the number of solvent lines and intensities are low. Atkinson, Phillips and other participants suggested these means for minimizing solvent effects. A solvent which does not have any of these limitations is liquid Xenon because it does not generate Raman lines, is transparent throughout the UV to IR range and does not fluoresce was suggested by Rentzepis. However he emphasized that Xenon does not dissolve highly polar molecules.

The rejection of fluorescence which is practically inherent in most if not all resonance Raman experiments was discussed in great detail by M. Delahay who proposes a simple modified interferometer as a means for its elimination at any wavelength as long as the fluorescence lifetime is much longer than the excitation pulse and hence the Raman lines. Should the lifetime of fluorescence be approximately the same as the excitation pulse i.e. stimulated fluorescence emitted at the wavelength of the Raman no line practical means was presented for their separation.

The lectures presented may be divided into two sections. 1) chemical processes presented in the morning and 2) biological in the afternoon. The resonance Raman spectra of several polyenes were discussed in detail with the emphases on the detection and understanding of molecular transformation especially cis-trans isomerization. The importance of excited state symmetry and structure was found

to be significant and presents complications is the interpretation of the results since little is known concerning excited state electronic and vibrational structure. Two presentations in the afternoon on bacteriorhodopsin are closely connected with the polyenes since the bacteriorhodopsin chromophore consists of a protonated shift base polyene. It is of interest to note the rather perplexing data by Storbrucker a) The N-H and N-D bonds of the Schiff base are strongly bonded and yet b) the N-H and N-D bonds seem to disappear by pumping out the water solvent. It seems that either the Paman vibrational assignment is wrong or something new in science takes place.

Inverse Raman processes, presently with time resolution in the microsecond range were discussed, it was suggested that this techniques may be sensitive for studying the structure of small molecules and eventually maybe extended to the picosecond range. The importance of utilizing excited state absorption, emission and Raman spectroscopy was displayed in the presentation of picosecond time resolved data for the rather detail investigation of photodissociation of haloxromatics. In this picosecond study all excited states and metastable species and radicals were identified by their spectra. Kinetics of formation and decay were directly monitored from ~ 1 psec to 50 nanoseconds. Picosecond Raman were also shown for the metastable C-X

bonds.

The general impression from this meeting was that the field of time resolved nanosecond Raman in the resonance mode is quite established experimentally although there still difficulty in interpretation of the results mostly due to our lack of a data bank of excited state symmetry. Picosecond Raman is in its early infancy however, there is strong interest in this field and as more investigator become active in it is expected that many of the experimental difficulties will be resolved and this field will become a very influential means for understanding the dynamics and structure of short lived states and intermediates.

Conferences such as this one at the Royal institution provide, in my opinion a most desirable avenue for presenting and discussing the present status of a field. It is a vehicle for integrating dispersed knowledge and leads to the path to be followed for the development of new fields of science in a well thought, orderly and prudent manner.

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